

## IV. Scintillation Detectors

Sources:

J.B. Birks, *The Theory and Practice of Scintillation Counting*,  
New York, 1964

G.F. Knoll, *Radiation Detection and Measurement*,  
New York, 1989

S.E. Derenzo, *Scintillation Counters, Photodetectors and  
Radiation Spectroscopy*, IEEE Short Course *Radiation  
Detection and Measurement*, 1997 Nuclear Science Symp.

- Incident particles or photons excite atoms or molecules in the scintillating medium.
- Excited states decay under emission of photons, which are detected and converted into electric signals.

### 1. Scintillation materials

Both organic and inorganic materials,

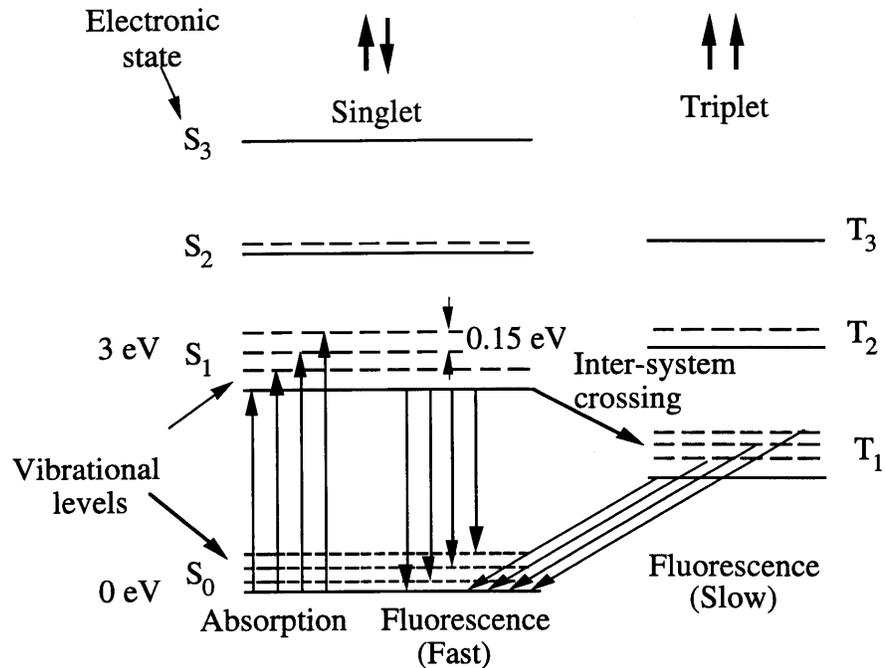
can be solid, liquid or gaseous

a) organic scintillators (e.g. plastics)

states of interest are energy levels of individual  
molecules, i.e. no interactions with neighbors

⇒ excitation and emission spectra practically the same  
whether in solid, liquid or gaseous state.

## Typical energy levels (from Birks, as redrawn by Derenzo)



- At room temperature practically all electrons in ground state.  
(since energy of  $S_1$  states  $\gg$  0.025 eV)
- Incident radiation populates  $S_1$  states  
  
vibrational levels within  $S_1$  band decay radiation-less to  $S_1$  base state, which in turn decays under emission of light to the  $S_0$  band.
- $S_1$  can also decay to adjacent triplet levels.  
Since their energy is significantly lower, the decay time is much longer.

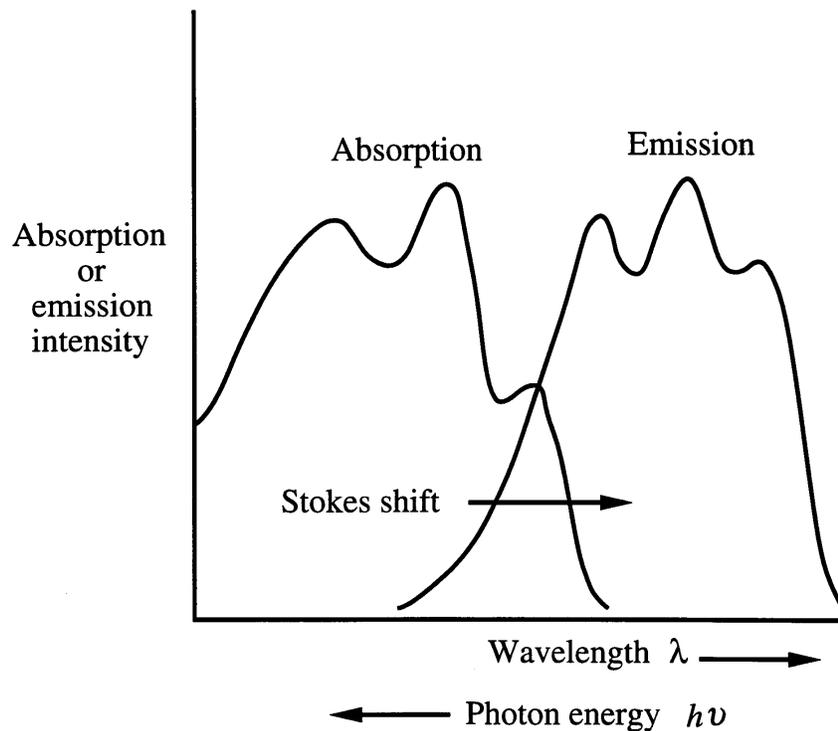
## Why isn't emitted light re-absorbed?

If emission and absorption occur at the same wavelengths, most emitted photons would be absorbed within a short distance,

⇒ poor light output from large volume scintillators

Since excitation goes to higher vibrational states in the  $S_1$  band, whereas decay goes from the base  $S_1$  state, the emission spectrum is shifted to lower energies (longer wavelengths).

⇒ only small overlap of emission and absorption spectra



## Time dependence of emitted light

a) non-radiative transfer of energy from vibrational states to fluorescent state

typical time: 0.2 – 0.4 ns

b) decay of fluorescent state

typical time: 1 – 3 ns

⇒ rise with time constant  $\tau_r$

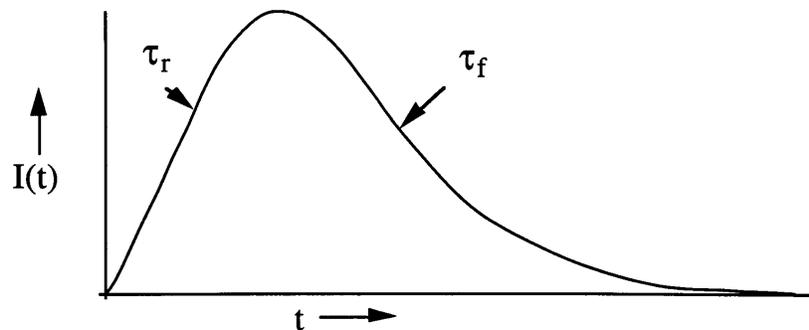
$$I(t) \propto 1 - e^{-t/\tau_r}$$

fall with time constant  $\tau_f$

$$I(t) \propto e^{-t/\tau_f}$$

total pulse shape

$$I(t) = I_0 (e^{-t/\tau_f} - e^{-t/\tau_r})$$



Rise time usually increased substantially by subsequent components in system and variations in path length in large scintillators.

## Properties of some typical organic scintillators

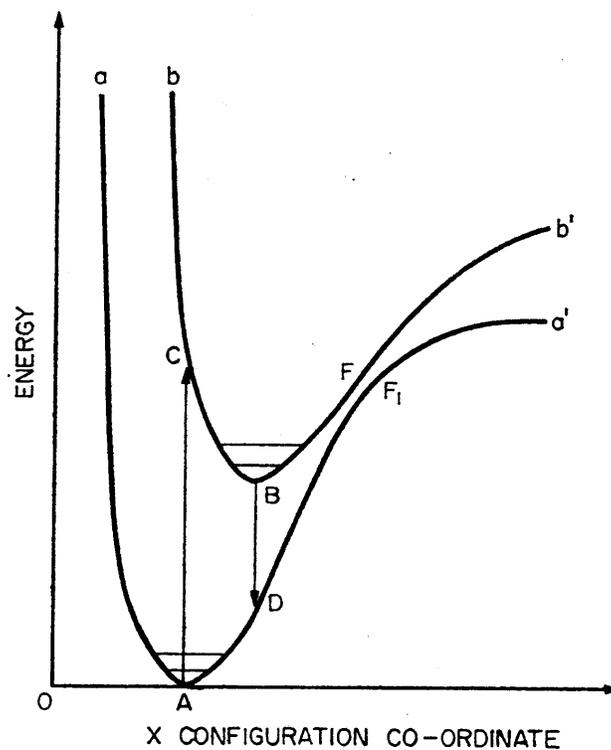
Material	State	$\lambda_{\max}$ [nm]	$\tau_f$ [ns]	$\rho$ [g/cm <sup>3</sup> ]	photons/MeV
Anthracene	crystal	447	30	1.25	$1.6 \cdot 10^4$
Pilot U	plastic	391	1.4	1.03	$1.0 \cdot 10^4$
NE104	plastic	406	1.8	1.03	$1.0 \cdot 10^4$
NE102	liquid	425	2.6	1.51	$1.2 \cdot 10^4$

Pilot U, NE104 and NE102 are manufacturers' designations

## Luminescence vs Quenching

Plot energy levels in configuration space, i.e. any set of coordinates describing the configuration of a molecule.

In general, configuration space is multi-dimensional, but one example is the interatomic distance, which may change as a molecule is excited. Since electronic transitions proceed much more rapidly than rearrangement of the nuclei (or the lattice in a crystal), they are represented by vertical lines in configuration space.

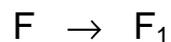


(from Birks)

Excitation:	$A \rightarrow C$	(very fast)
thermal equilibration:	$C \rightarrow B$	( $\sim 10^5$ longer)
Photon emission:	$B \rightarrow D$	
thermal equilibration:	$D \rightarrow A$	

The energy released in the transitions  $C \rightarrow B$  and  $D \rightarrow A$  is thermally dissipated as molecular or lattice vibrations.

If an excited electron reaches F (depending on population of states in minimum B), the transition



can proceed without emission of a photon.

In this case the sequence following an excitation is

Excitation:  $A \rightarrow C$

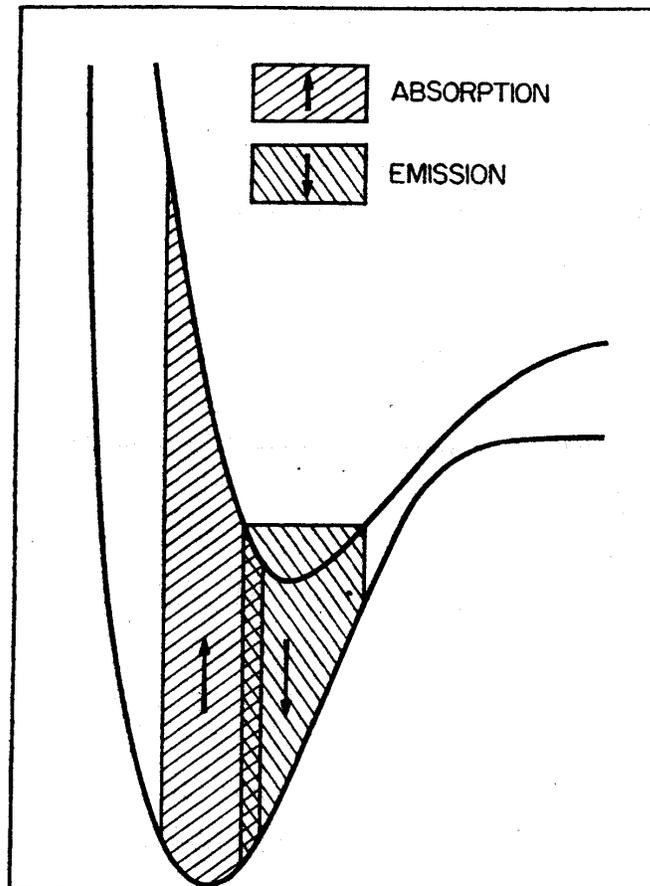
Decay to ground state:  $F \rightarrow F_1$

thermal equilibration:  $F_1 \rightarrow A$

This mode of decay without emission of a photon is called quenching, as it competes with the scintillation process and reduces the intensity of the emitted light.

In some crystals, the proximity region F-F<sub>1</sub> is very close to the minimum of the excited state. These crystals are heavily quenched.

## Overlap of absorption and emission spectra



(from Birks)

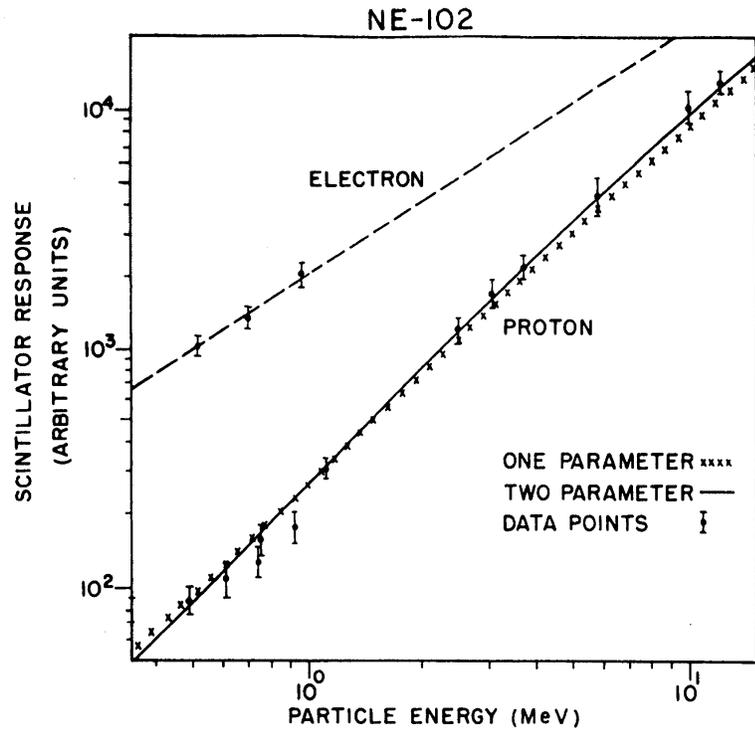
Thermal excitation populates states near minima.  
Widths of absorption and emission spectra depend on the density of states in the respective minima A and B.

A and B must be sufficiently separated to yield adequate Stokes shift.

At high temperatures the absorption and emission bands broaden, increasing the overlap and the fraction of luminescence photons lost to self-absorption.

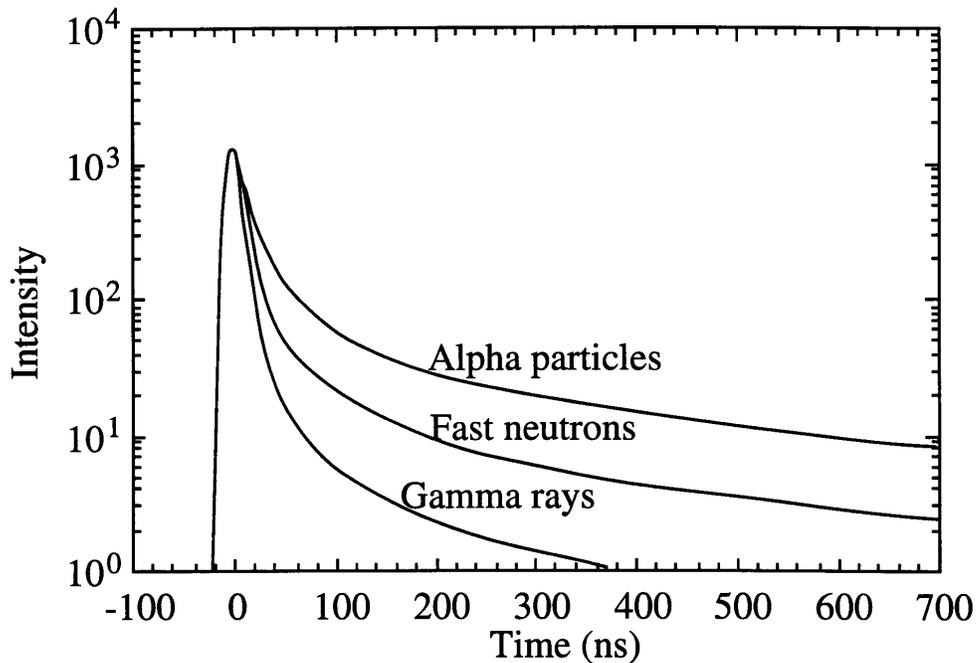
⇒ reduction in light output with increasing temperature

Both the light output and the decay time of organic scintillators depend on the ionization density.



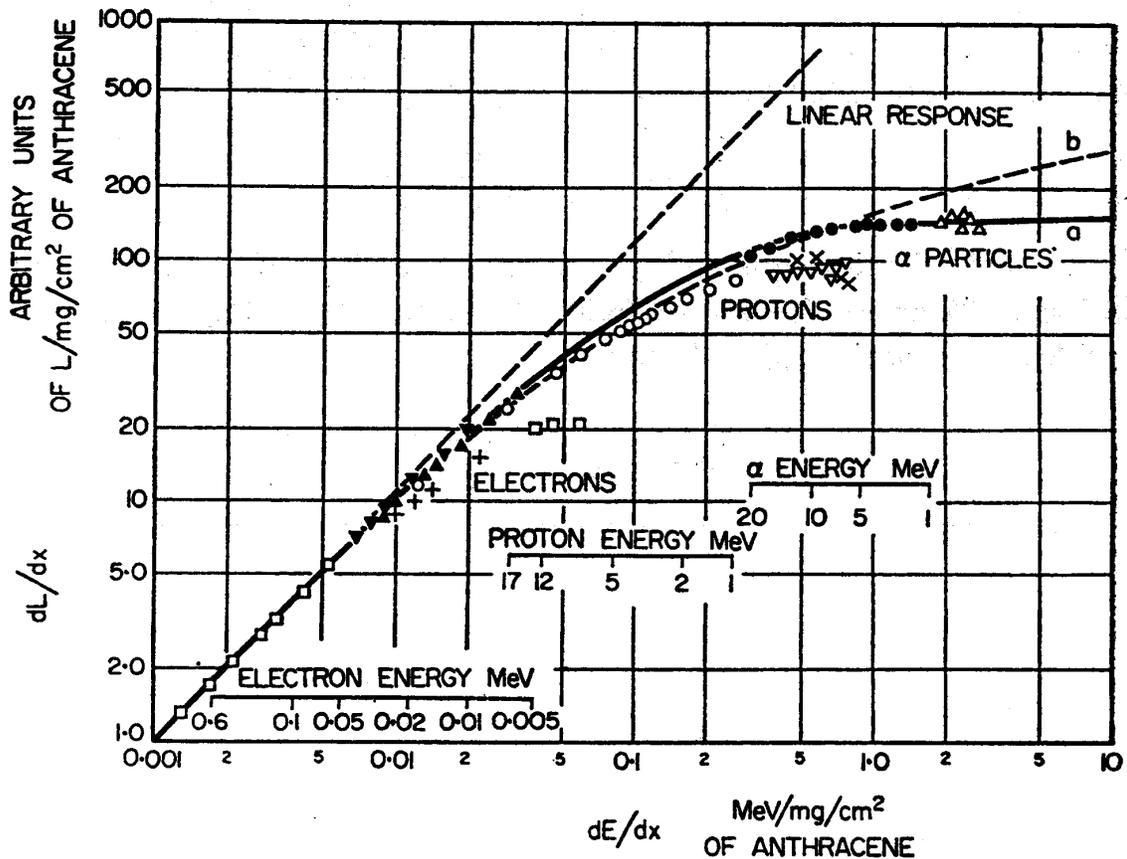
(Craun and Smith)

Decay time in stilbene for various particles



(from Bollinger and Thomas)

Variation of specific fluorescence  $dL/dx$  in anthracene with specific energy loss  $dE/dx$



(Brooks, from Birks)

The fact that data for both weakly and heavily ionizing particles lie on the same curve of light output vs.  $dE/dx$ , shows that the reduction in light output depends on ionization density.

Interpretation:

For lightly ionizing particles, e.g. electrons, the spacing between successive ionizations is several molecular distances, so the interaction between ionization sites is negligible.

Heavily ionizing particles lead to overlapping excitations, which tends to increase quenching.

## Birk's Rule

For an ideal scintillator and low ionization density

Luminescence  $\propto$  Energy dissipated in scintillator

$$L = SE$$

or, in differential form

$$\frac{dL}{dr} = S \frac{dE}{dr}$$

The specific density of ionized and excited molecules along the particle track is

$$B \frac{dE}{dr}$$

Assume that a portion of the primary excitation is lost at high ionization density (ionization quenching) and introduce a quenching parameter  $k$ . Then

$$\frac{dL}{dr} = \frac{S \frac{dE}{dr}}{1 + kB \frac{dE}{dr}}$$

For small  $dE/dr$  this yields the luminescence yield postulated above.

For large  $dE/dr$  the specific luminescence saturates, as indicated by the data.

$$\frac{dL}{dr} = \frac{S}{kB} = \text{const}$$

The dependence of decay time on ionization density can be used for particle identification.

For example, by utilizing a pulse shaping network that makes the timing of the output pulse dependent on decay time, the particle distribution is transformed into a time distribution that can be digitized directly.

### Example: n- $\gamma$ discrimination

P. Sperr, H. Spieler, M.R. Maier, NIM **116**(1974)55

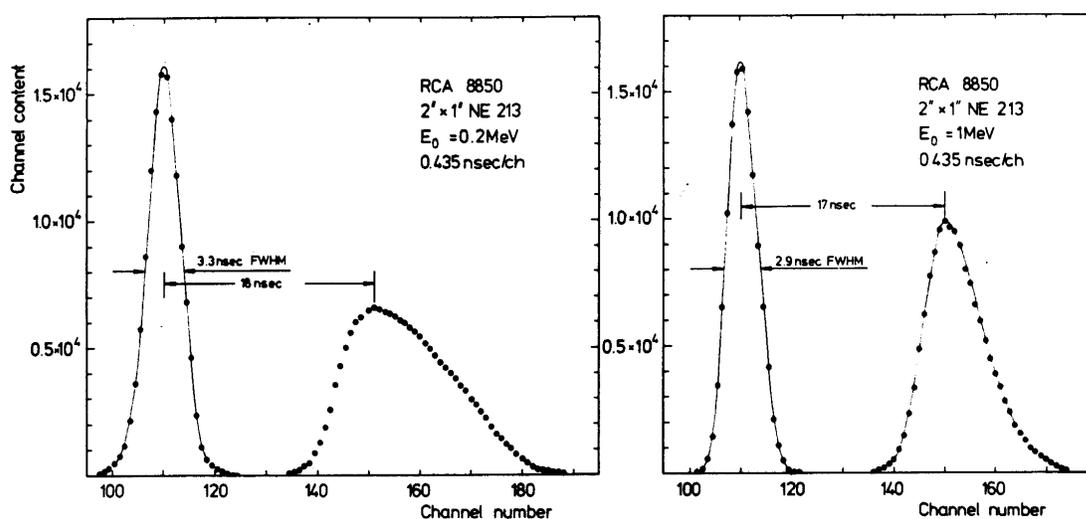


Fig. 5. Neutron-gamma timing distributions with a small scintillator (2" diam.  $\times$  1") for two threshold energies.

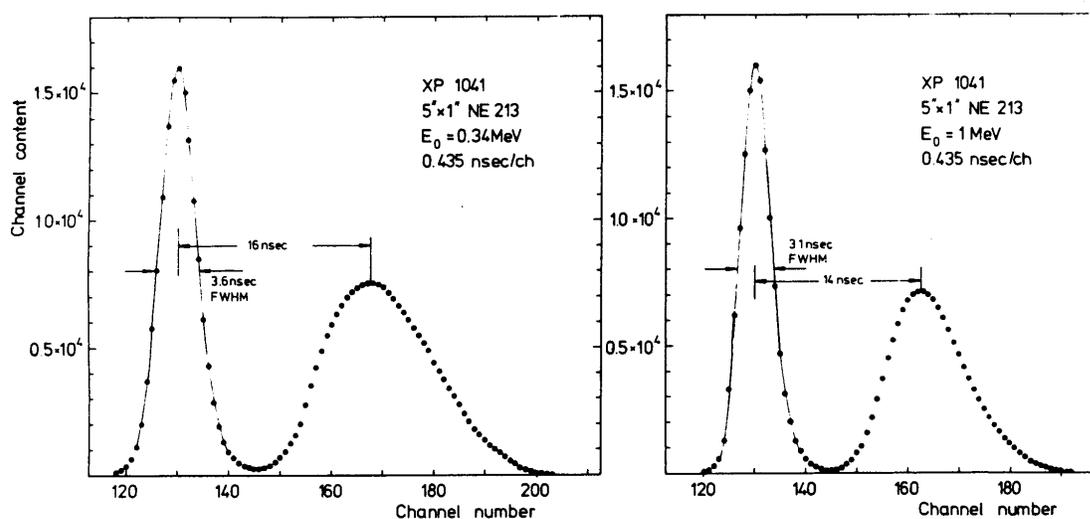
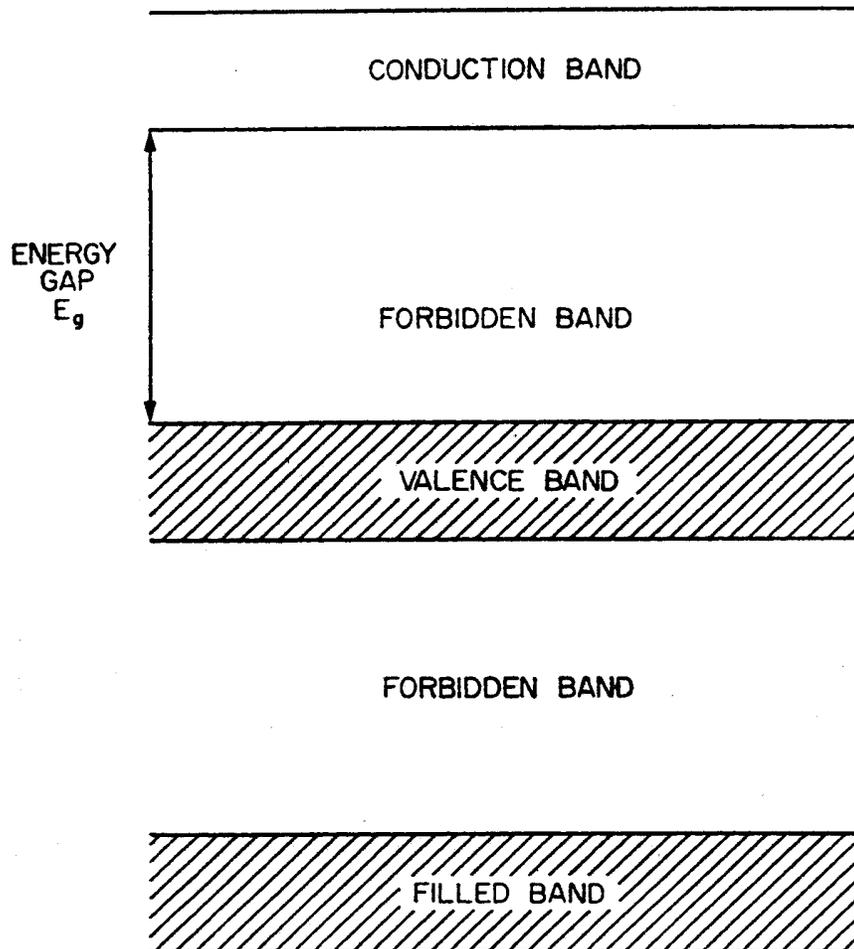


Fig. 6. Neutron-gamma timing distributions with a 5" diam.  $\times$  1" scintillator for two threshold energies.

# Inorganic Scintillators

Band structure in inorganic crystals



If forbidden band  $\gg kT$ , no electrons in conduction band.

$\Rightarrow$  Insulator

Radiation excites electron from valence into conduction band, forming an electron-hole pair.

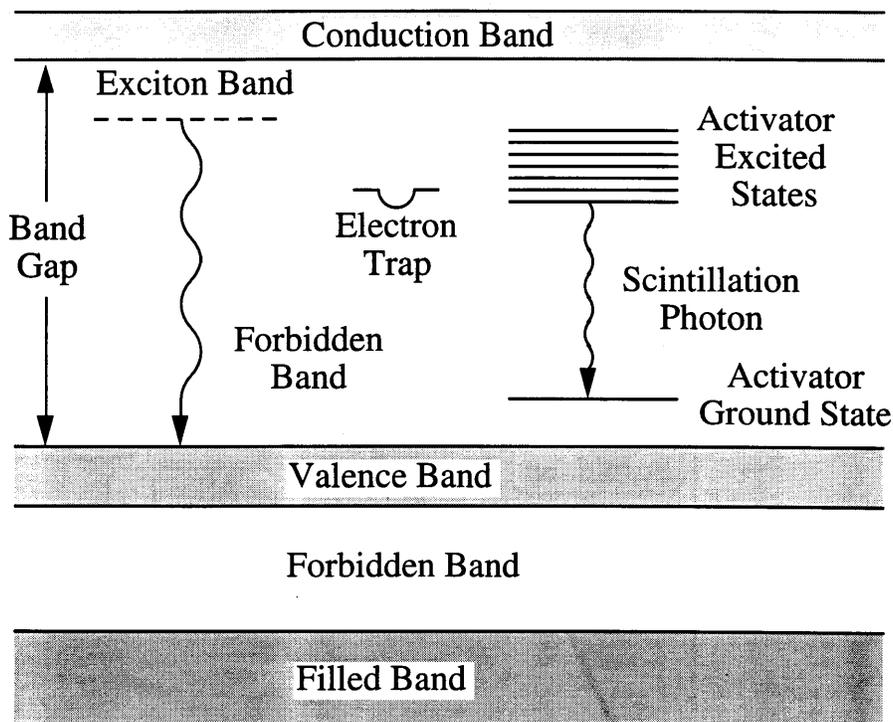
Electrons in conduction band and holes in valence band can move freely throughout crystal.

For light emission, one must introduce states into the forbidden band, so that

$$E_{\text{emission}} < E_g$$

Three mechanisms:

- a) excitons (bound electron-hole pair)
- b) defects (interstitial atoms, for example induced by heat treatment)
- c) activators  
very dilute concentrations of specific atoms can greatly increase light output



(from Derenzo)

Examples:

cooled NaI:  $h^+ + e^- \rightarrow \text{exciton} \rightarrow \text{phonons} + \text{photon}$

NaI(Tl):  $h^+ + \text{Tl}^+ \rightarrow \text{Tl}^{2+}$        $e^- + \text{Tl}^{2+} \rightarrow (\text{Tl}^+)^*$   
 $e^- + \text{Tl}^+ \rightarrow \text{Tl}^0$        $h^+ + \text{Tl}^0 \rightarrow (\text{Tl}^+)^*$   
 $(\text{Tl}^+)^* \rightarrow \text{Tl}^+ + \text{phonons} + \text{photon}$

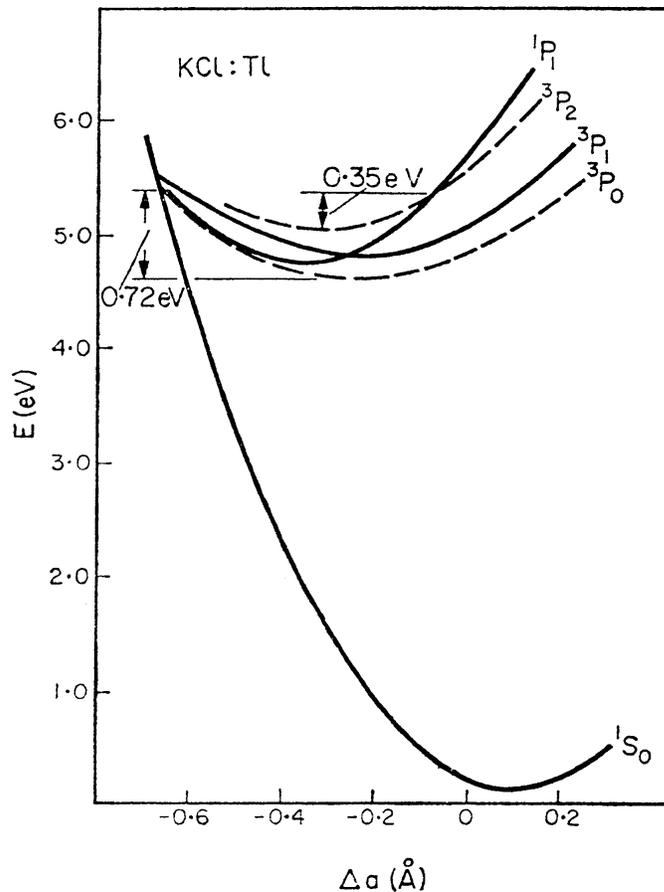
## Summary of practical inorganic scintillator materials (from Derenzo)

Material	Form	$\lambda_{\max}$ (nm)	$\tau_f$ (ns)	$\rho$ (g/cm <sup>3</sup> )	Photons per MeV
NaI(Tl) (20°C)	crystal	415	230	3.67	38,000
pure NaI (-196°C)	crystal	303	60	3.67	76,000
Bi <sub>4</sub> Ge <sub>3</sub> O <sub>12</sub> (20°C)	crystal	480	300	7.13	8,200
Bi <sub>4</sub> Ge <sub>3</sub> O <sub>12</sub> (-100°C)	crystal	480	2000	7.13	24,000
CsI(Na)	crystal	420	630	4.51	39,000
CsI(Tl)	crystal	540	800	4.51	60,000
CsI (pure)	crystal	315	16	4.51	2,300
CsF	crystal	390	2	4.64	2,500
BaF <sub>2</sub> (slow)	crystal	310	630	4.9	10,000
BaF <sub>2</sub> (fast)	crystal	220	0.8	4.9	1,800
Gd <sub>2</sub> SiO <sub>5</sub> (Ce)	crystal	440	60	6.71	10,000
CdWO <sub>4</sub>	crystal	530	15000	7.9	7,000
CaWO <sub>4</sub>	crystal	430	6000	6.1	6,000
CeF <sub>3</sub>	crystal	340	27	6.16	4,400
PbWO <sub>4</sub>	crystal	460	2, 10, 38	8.2	500
Lu <sub>2</sub> SiO <sub>5</sub> (Ce)	crystal	420	40	7.4	30,000
YAlO <sub>3</sub> (Ce)	crystal	390	31	5.35	19,700
Y <sub>2</sub> SiO <sub>5</sub> (Ce)	crystal	420	70	2.70	45,000

Note the wide range of decay times  $\tau_f$ , from 0.8 ns in BaF<sub>2</sub> to 15  $\mu$ s in CdWO<sub>4</sub>.

Some materials also show multiple emissions (BaF<sub>2</sub>, PbWO<sub>4</sub>).

Scintillators with Tl, Bi, etc. decay slowly because direct transitions from the lowest-lying excited state to the ground state are forbidden:



(from Birks)

Initially, the lowest-lying state ( $3P_0$ ) is populated.

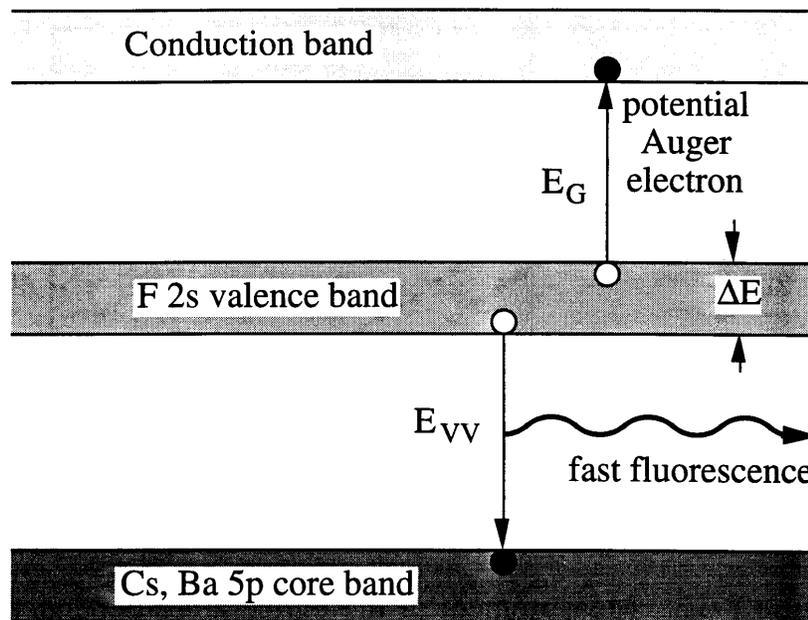
Transitions between the triplet states  $3P_0$ ,  $3P_2$  and the  $1S_0$  ground state are forbidden ( $\Delta S = 1$ ,  $\Delta j = 0, 2$ ).

The transition from  $3P_1$  to the ground state  $1S_0$  is allowed.

The transition from  $1P_1$  to the ground state  $1S_0$  is allowed, but weak, as the energy difference from the  $1P_1$  potential minimum to the ground state is substantially less than for the  $3P_1 - 1S_0$  transition.

The dominant emission is due to the  $3P_1 - 1S_0$  transition, which is populated slowly by thermal activation from the  $3P_0$  state.

The very fast transitions in BaF<sub>2</sub> and CsF are due to an intermediate transition between the valence and core bands.



(from Derenzo)

$$E_{vv} < E_g$$

fast fluorescence

$$E_{vv} > E_g$$

emission of Auger electron

(energy released in the transition from the valence to the core band does not go into photon emission, but into emission of an electron to the conduction band)

Competition between photon emission and Auger effect narrows the range of scintillators with fast decays:

If  $E_{vv}$  is low: longer wavelength emission, longer decay time

If  $E_{vv}$  is high: Auger emission, no scintillation light